

Study of Electrical Characteristics of Some Organic Semiconductors for Light-emitting and Photovoltaic devices

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ABSTRACT

Organic semiconductors have recently gained much attention due to their potential applications such as organic light emitting diodes (OLEDs), organic field effect transistors (OFETs) and organic photovoltaics. In this paper we concentrate on electrical properties of materials potentially applicable as elements of electronic devices: thin film field-effect transistors, and light-emitting and photovoltaic devices.

Keywords: organic semiconductor, charge transport mechanisms.

1. INTRODUCTION

Organic semiconductors are at the top of the considerable interest due to the possibility to make electronic devices on flexible materials (for example plastic or paper) using cheap technological processes such as spin coating and printing. In order to improve efficiency of an organic semiconductor device, it is important to have a comprehensive knowledge of electrical characteristics of organic semiconductors. An organic semiconductor is an organic material that has semiconductor properties. Semiconductivity is exhibited by single molecules, short chain (oligomers) and organic polymers. Semiconducting small

molecules (aromatic hydrocarbons) include the polycyclic aromatic compounds pentacene, anthracene, and rubrene. Examples of polymeric semiconductors are poly (3-hexylthiophene), poly (p- phenylene vinylene), as well as polyacetylene and its derivatives.

There are two major overlapping classes of organic semiconductors: organic charge-transfer complexes and various linear-backbone conductive polymers derived from polyacetylene, such as polyacetylene itself, polypyrrole, and polyaniline. Charge- transfer complexes often exhibit similar conduction mechanisms to inorganic semiconductors, at least locally. Such mechanisms arise from the presence of

hole and electron conduction layers separated by a band gap. As with inorganic amorphous semiconductors, tunneling, localized states, mobility gaps, and phonon-assisted hopping also contribute to conduction, particularly in polyacetylenes. Like inorganic semiconductors, organic semiconductors can be doped. Organic semiconductors susceptible to doping polyaniline (PANI) and poly (ethylenedioxythiophene) PEDOT: PSS are also known as organic metals¹⁶.

Typical carriers in organic semiconductors are holes and electrons in π -electrons. Almost all organic solids are insulators. But when their constituent molecules have π -conjugate systems, electrons can move via π -electron cloud overlaps. Polycyclic aromatic hydrocarbons and phthalocyanine salt crystals are examples of this type of organic semiconductor.

In charge-transfer complexes even unpaired electrons may be stable. Such unpaired electrons can function as current carriers. This type of semiconductor is also obtained by pairing an electron donor molecule and an electron acceptor molecule¹⁶.

2. LITERATURE REVIEW

Initially, the research on organic materials was concentrated on single crystals of organic materials, which can have mobilities of a few cm^2/Vsec at room temperature. However, for practical applications, thin film organic semiconductors with disordered morphology are required because the operating voltages for crystals were quite high (~ 100 V). The work on thin film devices have been started after

the realization of efficient bilayer OLED. Since then, OLEDs based on small conjugated molecules were intensively studied, and improved, which led to high brightness, long term stability and excellent color purity¹⁻³.

Organic semiconductors are of particular scientific and industrial interest because of their advantages such as availability, low cost, color tunability and the ability to grow on flexible substrates. Due to their several advantages, the application area of OLEDs is very broad. These applications include full color display⁴, solid state lighting, and organic memory devices.

3. MATERIALS

3.1 Molecules

Table 1 presents chemical structures of some molecules and groups of molecules whose properties will be analysed in this paper.

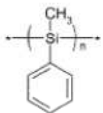
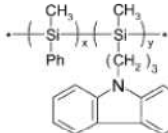
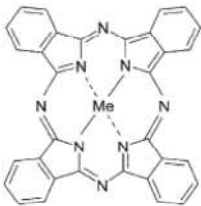
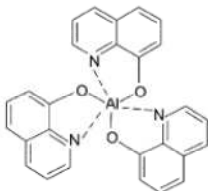
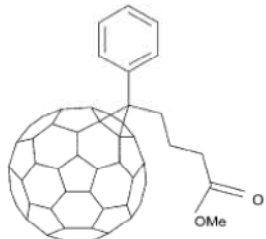
Table 1 contains formulae of a few groups of prototypic electroactive polymers. These polymers and their derivatives are commonly used in experimental research, and are likely to emerge in commercial devices. When undoped, all of them are wide band-gap semiconductors. Thus the thermal generation of carriers is possible only upon doping; using the terminology borrowed from inorganic semiconductors, they are extrinsic semiconductors. For many applications it would be desirable to have an intrinsically conducting polymer, *i.e.*, a conjugated polymer with a narrow energy gap. Various methods of manipulating the band gap in conjugated polymers have been

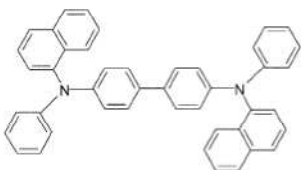
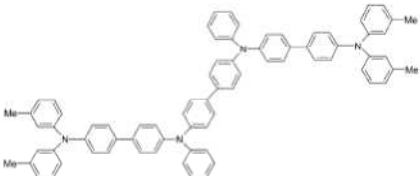
thoroughly discussed in the recent review by van Mullekom *et al.*,⁵ we will therefore only briefly address here two approaches aimed at minimizing the band gap.

Table 1 Molecular structures, names and acronyms of common molecules described in this article; only the structures of a few prototypic materials have been shown

Structure	Name and acronym used in this paper
	Polycyclic aromatic hydrocarbons. Acenes: $n = 1$: anthracene (Ac) $n = 2$: tetracene (Tc) $n = 3$: pentacene (Pc) $n \rightarrow \infty$: polyacene (PAC)
	Perylene
	Oligo- and poly- <i>p</i> -phenylenes $n = 2$: <i>p</i> -quaterphenyl (4P) $n = 3$: <i>p</i> -quinquephenyl (5P) $n = 4$: <i>p</i> -sexiphenyl; <i>p</i> -hexaphenyl (6P) $n \rightarrow \infty$: poly- <i>p</i> -phenylene (PPP)
	Polyfluorene (PF)
	Ladder-type poly- <i>p</i> -phenylene (LPPP)
	Oligo- and poly- <i>p</i> -phenylenevinylenes $n = 1$: <i>p</i> -distyrylbenzene $n \rightarrow \infty$: poly- <i>p</i> -phenylenevinylene (PPV)
	Poly(2-methoxy-5-(2'-ethylhexoxy)- <i>p</i> -phenylene vinylene) (MEH-PPV)
	Oligo- and poly- α -thiophenes $n = 2$: α -quaterthiophene (4T) $n = 3$: α -quinquethiophene (5T) $n = 4$: α -sexithiophene; α -hexathiophene (6T) $n = 6$: α -octithiophene (8T) $n \rightarrow \infty$: polythiophene (PT)
	Polyisothianaphthene (PITN)

Table 1 (Contd.)

Structure	Name and acronym used in this paper
	Polysilanes (polysilylenes): Poly(methylphenylsilane) (PMPSi)
	PSiK
	Metal phthalocyanine (MePc)
	Tris(8-hydroxyquinolino)aluminium (Alq ₃)
	Fullerenes: C ₆₀
	1-(3-Methoxycarbonyl)propyl-1,1-phenyl-(6,6)C ₆₁ (PCBM)
	Amines: N,N'-diphenyl-bis(3-methylphenyl)- (1,1'-biphenyl)-4,4'-diamine (TPD);

Structure	Name and acronym used in this paper
	<i>N,N'</i> -diphenyl-bis(1-naphthyl)(1,1'-biphenyl)-4,4'-diamine (NPB);
	<i>N,N'</i> -diphenyl- <i>N,N'</i> -bis(di(3-methylphenyl)aminobiphenyl)benzidine (TPTE)

The band gap can be abridged if the bond-length alternation is cancelled. The best recognized example of such a polymer is a polyisothianaphthene (PITN), which can be considered as a derivative of polythiophene (PT). In contrast to PT, in which the quinoidal form is energetically not favorable because of the loss of aromaticity, in PITN the loss of aromaticity in the heteroatomic ring is compensated by gaining aromaticity in the fused six-member ring. This reduces the energy gap from *ca.* 2 eV for PT to *ca.* 1 eV for PITN. Another class of polymers with reduced bond-length alternation are ladder polymers. Polyacene is the simplest example of the ladder polymer which, according to theoretical considerations, should exhibit a metallic conductivity. Unfortunately, its synthesis is not simple at all. It should be mentioned here that in other ladder-type polymers, LPPP, widely used as electroluminescent materials, the bond-length alternation effect remains and some reduction in the gap width is only due to the rigid backbone with planar

neighbouring rings.

3.2 Morphology

Morphology (materials science), the study of shape, size, texture and phase distribution of physical objects.

Several reports on the escalation of single crystals of oligothiophenes have been published in recent years, the interest being focused on long oligomers (4T and longer molecules). Thin crystalline flakes of unsubstituted thiophenes have been grown from the vapour phase, although melt-grown crystals have also been studied. The dense packing of the molecules results in most cases in their herring-bone arrangement, the long axes of all molecules in the unit cell assuming a parallel (or nearly parallel) orientation. The growth conditions may play some role in the molecular packing, resulting in obtaining different crystal modifications, as has been found in 4T and 6T (so-called 'low-temperature' and 'high temperature' polymorphs). Substitution at the ends or at the

sides of the molecules can modify the π orbital systems hence influencing the packing details and the overlap of molecular charge transport.

Table 2 Ionization energies, electron affinities and energies of HOMO and LUMO levels in a few organic solids. The values labelled I_e and A_e have been determined from photoemission experiments and reported values of band gaps, those labelled HOMO and LUMO are given as reported in the papers quoted without specifying the method or conditions of determination. Some values given in the table have been compiled from results reported in the quoted references. See the references for the original literature. Energies are given to within 0.1 eV

Molecule	I_e /eV	A_e /eV	HOMO/eV	LUMO/eV	Ref.
Naphthalene	6.4				30
	6.8	1.8			1
Anthracene	5.9				2,31
		1.8			
	5.7				30
	5.8	2.0			1
Tetracene	5.3	2.2			6
	5.1				30
	5.4	2.4			1
Pentacene	5.0	2.5			6
	4.9				30
	5.1	2.9			1
Perylene	5.2				30
	5.4	2.5			1
6P	5.8–6.0	2.7–2.9			32
C ₆₀	6.9	3.4			33
			6.4	4.5	34
			6.2	4.5	35
			6.1	3.7	36
TPD	5.9				37
			5.5	2.3	38
			5.8	2.6	39
NPD	6.4				37
			5.4	2.4	38
			5.7	2.6	39
BCP			6.1	2.4	40
CuPc	5.0				41
			5.3	3.6	42
			5.2	3.5	35
ZnPc	5.2				41
Alq ₃	6.3				43
	5.7	3.0	5.7	3.0	42
			5.8	3.1	38
PPy			4.0	1.6	44
PMeT			4.9	3.0	44
PHT			5.2	3.2	44
MEH-PPV			4.9	2.8	34
			5.0	2.8	36
PMPSi			5.3	1.8	45
PVK			6.1	2.1	46

4. ENERGY LEVELS, CONTACTS

Electrical properties of molecular materials are determined, among other factors, by energies of levels (bands) allowed for electrons and holes in these

materials. In perfect molecular crystals, the transport occurs in narrow energy bands; in disordered (poly-crystalline or amorphous) materials, the bands split into a manifold of local states. If the vacuum level is taken as reference, then the energies of the electron and hole levels are, respectively, the electron affinity of molecules and their ionization energy in the solid state. The electrical energy gap is then the difference of the two latter parameters, being a measure of the energy difference between the energies of levels allowed for non-interacting electrons and holes.

At this instant, a terminology-related problem should be invoked. The energy gap is often identified with the optical (HOMO-LUMO) gap taken directly from spectroscopic measurements. Moreover, in some cases the energies of the HOMO and LUMO levels used to calculate the gap are taken from quantum-chemical calculations performed for isolated molecules. The former procedure neglects the electron-hole interactions and is likely to underestimate the gap by some 0.5 eV. The latter approach neglects the electrostatic interaction of a charge carrier with the polarizable environment introducing an important error, often exceeding 1 eV. Furthermore, approximate values of the HOMO and LUMO energies are often deduced from electrochemical measurements of the first oxidation and reduction potentials. This method requires a proper calibration.

Typically, the solid-state ionization energies of various organic solids of interest range between 4 and 6 eV, their gaps - between 2 and 4 eV (*cf.* Table 2). Within groups of homologues, the

ionization energies and the gaps usually decrease with increasing size of the molecule.

This relation is illustrated in Table 2 with acenes. A similar relation is observed in other groups of compounds: for example, the electron-hole gap in oligothiophenes decreases from 3.6 eV in 4T to 3.0 eV in 6T⁶.

The mainstream of typical non-substituted low-molecular weight materials (acenes, phenylenes, thiophenes, phthalocyanines, . . .) are p-type (hole-conducting) materials. A analogous circumstance is encountered in π -conjugated polymers: most of them are p-type semiconductors built of electron-releasing macromolecules consisting of electron-rich subunits (like thiophene or pyrrole). Stable materials with electro accepting properties (n-type materials) are required in all important electronic and opto-electronic applications: (bipolar transistors, photovoltaic devices, LEDs and FETs, . . .). So far, however, only a few devices with such materials have been studied due to a shortage of suitable acceptor polymers. In polymer LEDs, since p-type conjugated polymers are much better hole- than electron-transporting materials, the resulting charge imbalance limits EL efficiency. Additionally, polymers with high electron affinities would allow the use of aluminium as the cathode, which is much more stable than the often used at present low-work-function calcium electrode. In photovoltaic devices, preparation of composites of separated, but inter-penetrating phases of acceptor- and donor-type conjugated polymer should allow one to use the 'bulk heterojunction' concept in producing PV

cells. Some of such devices have already been produced, showing conversion efficiency similar to those obtained for polymer/fullerene systems. As in the heterojunction systems with fullerenes, MEH-PPV was used as a donor-type polymer, and the high electron affinity polymer was obtained by cyano substitution of MEH-PPV. All-polymer 'bulk heterojunctions' open new perspectives for PV devices. For example, it was shown that it is possible to create a bilayer junction with n- and p-type polymers interpenetrating each other. The use of organic materials in photovoltaic devices will be discussed below.

An palpable choice for low-molecular weight n-type materials are molecules with high electron affinity which, in most cases, means the use of molecules such as derivatives of oligo- and polythiophenes,⁷ oligo- and polyfluorenes,⁸ naphthalene-tetracarboxylic diimide (NTCDI)⁹ or phthalocyanines, cyano- or fluorine- substituted. One of the approaches for obtaining electron-transporting polymeric materials is to mix low molecular weight acceptors with inert polymer matrix. However, it is usually very difficult to obtain 'solid solutions' of acceptors molecularly dispersed in polymer matrix at concentrations sufficiently high to assure effective electron transport, since such systems are thermodynamically unstable and show a tendency to a phase separation. Especially at higher temperatures, inevitable during the fabrication procedure and also usually encountered by the working devices, the low molecular weight additives precipitate easily. This problem can be overcome, if electron-acceptor groups (like

aryl units with nitro- or cyano-groups), capable of n-doping and electron transporting properties are incorporated as side groups or in the polymer main chain. Many low molecular weight acceptors were used as building units to produce electron transporting polymers, such as: pyridine, oxadiazole, quinoline, quinoxaline, triazine, bithiazole and then were tested in PLED devices. It was found that these heterocyclic units act also as fluorophores emitting in most cases blue light, however usually with a poor quantum efficiency (QE)¹⁰.

Interfaces between organic solids and electrodes play a key role in nearly all processes determining the action of organic electronic devices, in particular, in OLEDs and in PV cells. A detailed discussion of these processes is clearly beyond the scope of this article, it will nevertheless be necessary to provide a brief description of basic factors influencing the behaviour of contacts and heterojunctions. The basic factor is the energetics of the interfaces, and in particular relations between the energy of electrons in the contact and the energies of the electron and/or hole transport levels. According to a 'textbook approach', an effective injection of electrons into an organic solid takes place from an electrode whose work function is close to or lower than the electron affinity of the molecules forming the solid, whereas the hole injection occurs from an electrode whose work function is close to or higher than the solid-state ionization energy of the material. This simple picture is usually obscured by the presence of surface effects of various kind¹¹ but nevertheless it can serve as a useful qualitative indication of injection ability of various electrodes.

5. PHOTOGENERATION OF CHARGE CARRIERS IN CONJUGATED POLYMERS

The mechanism of the most fundamental process involved in photoconductivity and photovoltaic phenomena in molecular crystals— photogeneration of charge carriers— is reasonably well understood in conjugated polymers, however, the carrier photogeneration has been controversial for a long time, in spite of an increasing number of experimental facts. The main point of the controversy is related to the strength of the photoexcited geminate electron–hole pair attraction as compared to the bandwidth, determining if the photoexcitations are strongly correlated Frenkel excitons or rather weakly bound excitons, allowing one to apply the band model with ultrafast charge photogeneration. In other words, the exciton binding energy determines whether conjugated polymers behave in this respect more like inorganic semiconductors or rather as other organic solids.¹².

The model assuming the band to band transition or ultrafast electron transfer from primary generated intra-chain excitons is based on the observation of a very fast appearance of charge carriers, well documented by several experiments with very high time resolution. Transient excited-state absorption spectroscopy experiments performed in the range of the infrared active vibrational (IRAV) modes in conjugated polymers (PPV and MEH-PPV) and in their composites (MEH-PPV/C60) showed that charge carriers are photogenerated in a time span shorter than 100 fs, with the quantum efficiency as high as ca.

0.1¹³. Since no correlation was found between the time evolution of the photoinduced IRAV signals and the exciton lifetime (ca. 300 ps), it was concluded that the charge carriers are photogenerated directly and not via the exciton annihilation. Pump–probe spectroscopy experiments for these polymers¹⁴. Showed a weak spectral dependence of charge generation quantum efficiency in the range from the absorption edge up to 6.2 eV. This result yielded a further argument for a direct photogeneration of charged polarons at times shorter than 100 fs, contrary to the ‘exciton models’ assuming indirect charge generation. Furthermore, it was argued that the dramatic increase of photocurrent for excitation energies above the onset of absorption in PPV derivatives reported in the literature (which had been explained by a large exciton binding energy) arises in fact from the external current generated by electron photoemission.

6. MOBILITY

6.1 Mobility measurements

Mobility is one of the essential parameters characterizing the transport of charges, hence numerous attempts have been reported aimed at determining this parameter, its dependence on temperature, electric field, and its relation to the morphology of samples. Although it is beyond the scope of this article to discuss details of the methods, it will be nevertheless useful to provide a brief description of the methods, commonly employed in determining the mobilities of charge carriers in organic solids. These are: (i) the time-of-flight (TOF) technique, (ii) the field-effect

transistor (FET) method. Other methods are used less frequently or yield less reliable values.

Conceptually, the TOF technique is the simplest method, based directly on the definition of the mobility. A thin sheet of charge carriers is created in the surface region of a plane-parallel sample provided with two electrodes (Fig. 1). The measurement then consists in determining the time necessary for the carriers to reach the counter electrode (transit time, t_{tr} – cf. Fig. 1b) under a given biasing voltage V . The drift mobility μ is calculated from the equation

$$\mu = \frac{L^2}{t_{tr} V} \quad (1)$$

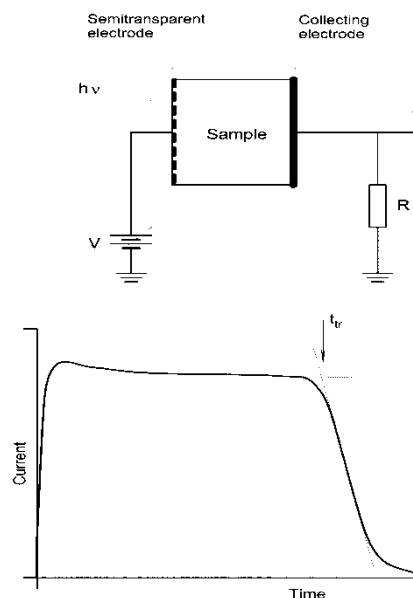


Fig. 1 TOF experiment. A scheme of the TOF apparatus (a), and a typical transient current as seen on the oscilloscope (b); t_{tr} is the transit time.

where L is the model thickness. The technique can be used for materials

exhibiting low dark conductivities. Moreover, the material under study should be either photoconducting or it should be provided with a suitable photoinjecting contact. The drift mobility so determined is an effective parameter which may be affected by traps (*vide infra*). In the limiting case, for a sample with deep traps whose trapping time is shorter than the transit time, the latter cannot be determined.

FET is a three-terminal device, schematically depicted in Fig. 2a. The experiment consists of measuring source-to-drain currents (I_d) as a function of the source-to-drain potential difference (U_d) at various gate voltages (U_g). A family of schematic current-voltage characteristics is shown in Fig. 2b. The mobility can be determined from either the equation describing the slope of the dependence of the source-to-drain current on the gate voltage, measured in the linear portion of the plot

$$\left(\frac{\partial I_d}{\partial U_g} \right)_{U_d = \text{const}} = \frac{W}{L} C_i \mu U_d \quad (2)$$

or from the dependence of the saturation current on the gate voltage.

$$I_{d,\text{sat}} = \frac{W}{2L} C_i \mu (U_g - U_0)^2 \quad (3)$$

In the above equations, W and L are channel width and length, respectively, C_i is the capacitance of the insulating layer (per unit length), and U_0 is the threshold voltage.

6.2 Mobilities in organic single crystals

For the discussion of the theories describing the transport of charge carriers in perfect molecular solids, the reader is

referred to a rich literature existing on the subject. For the purpose of the present discussion it is sufficient to mention that the theory predicts that the transport of charge carriers in perfect molecular solids occurs in narrow bands, the carriers being strongly coupled to lattice phonons. The experiments performed on single crystals, and in particular the temperature dependences of the mobilities, their anisotropies and relation to the crystal structure allow one to get an insight into microscopic phenomena controlling the transport of charge carriers and provide a reference for results obtained on less perfect samples. Experiments showed that the highest room-temperature mobilities measured in molecular single crystals are of the order of $10\text{--}101\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$, and increase to $101\text{--}102\text{ cm}^2\text{ V}^{-1}\text{ s}^{-1}$ at temperatures of the order of 10^1 .¹⁵

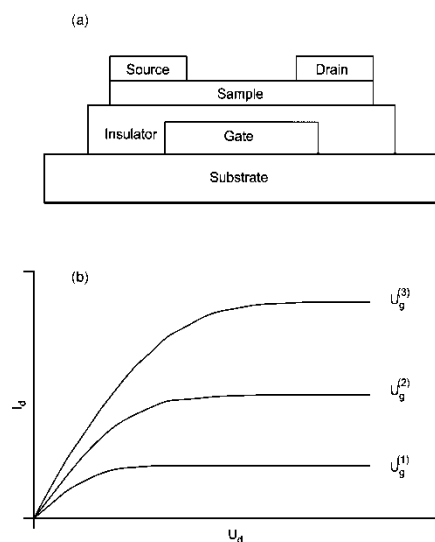


Fig. 2 FE experiment. A schematic view of a thin film FET (a), and a family of current-voltage characteristics (b). The curves have been plotted for different gate voltages ($U_g^{(1)} < U_g^{(2)} < U_g^{(3)}$).

A good example of such a behaviour is the temperature dependence of the drift mobility of holes and electrons in single crystals of polycyclic hydrocarbons. Careful measurements performed on single crystals demonstrate that the 'lattice' drift mobilities (μ_0) follow a T^{-n} dependence, with $0 < n < 3$ but mostly ranging between 1 and 1.5.

These experiments, however, can only be performed on samples of the highest chemical purity and controlled perfection, hence reliable results are scarce, and limited practically to poly-cyclic aromatic hydrocarbons. Early results obtained for single-crystalline samples have been listed by Iarl; in most cases, they have been obtained by the time-of-flight method. The mobilities determined for single crystals at biasing fields typical of this type of experiment (up to $\sim 10^3\text{--}10^4\text{ V cm}^{-1}$) are field-independent; at high fields (of the order of $10^4\text{--}10^5\text{ V cm}^{-1}$) the drift velocities of charge carriers tend to saturate thus resulting in a decrease of the apparent mobilities.

Doping of molecular solids with a small amount (of the order of $10^{-7}\text{--}10^{-4}\text{ mol mol}^{-1}$) of suitably chosen guest molecules may give rise to a dramatic reduction of the effective drift mobilities due to trapping of charge carriers. In the case of shallow traps controlling the transport, the effective mobility (μ_{eff}) is described by the equation first derived by Hoesterrey and Letson.

$$\mu_{\text{eff}} = \frac{\mu_0}{1 + x_g \exp(E_t / kT)} \quad (4)$$

where x_g is the mole fraction of the guest molecule, and E_t stands for the depth of the trap formed by the dopant (assumed monoenergetic). It follows from the equation

that at low temperatures the mobility should be temperature-activated with a slope determined by the trap depth, whereas at sufficiently high temperatures it should attain its 'lattice' value (μ_0) equal to that in a pure crystal.

The source of traps in molecular materials has been studied and methods of their determination have been developed in the seventies and eighties of the 20th century, and discussed in numerous monographs and reviews. In general, traps can be associated with either chemical impurities ('chemical traps') or with defects of crystal structure ('structural traps').

7. CONCLUSION

In this paper, we have discussed the major advances that have recently been achieved in the description of the parameters impacting semiconductors. Once again, the picture emerging in organic semiconductors appears to be more complex than in conventional inorganic semiconductors. In this paper we concentrated on electrical properties of materials potentially applicable as elements of electronic devices: thin film field-effect transistors, and light-emitting and photovoltaic devices.

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